

EPITAXIAL GROWTH
AND
DEEP LEVEL CHARACTERIZATION
OF
 $\text{GaAs}_{1-x}\text{P}_x$

PÄR OMLING



DEPARTMENT
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SWEDEN

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Abstract Evaluation of the growth and alloying mechanisms of the $\text{GaAs}_{1-x}\text{P}_x$ semiconductors using the new metalorganic vapour phase epitaxy (MOVPE) technique is performed. The material has been characterized optically and electrically. A method for tailor growth of material for systematical studies of deep levels, of which many are poorly understood, is presented. The following characterization techniques have been used: SEM including microprobe analysis, photo-luminescence, injection-luminescence, absorption, Hall effect measurements, and different forms of junction space charge techniques such as DLTS and phot capacitance measurements for deep level investigations. Successful epitaxial growth of the full ternary alloy system $\text{GaAs}_{1-x}\text{P}_x$ is presented. New experimental and analysis approaches are presented for the extraction of crucial data for vapour phase and surface chemistry mechanisms governing the solid phase composition of alloys. The grown material is found to have optical and electrical quality compatible with device requirements. From a study of GaAs:Fe it has been possible to determine the position of the deep level in the band gap and to correlate internal transitions with details in the photo-ionization spectrum. The deep donor EL2 has been studied in GaAs, and also in $\text{GaAs}_{1-x}\text{P}_x$ where it has been observed for the first time. The formation of the EL2-level is found to be limited by the V_{Ga} concentration. The phot capacitance quenching effect of EL2 previously observed in GaAs has been studied in the $\text{GaAs}_{1-x}\text{P}_x$ alloy system and optical and thermal properties of the metastable state have been determined. A previously unknown temperature dependence of the spectral distribution of the transition from the normal state to the metastable state in GaAs is reported and a possible explanation is discussed. In work on DLTS evaluation of nonexponential transients in semiconductor alloys it was found that DLTS-deduced activation energies and thermal emission rates are, indeed, relevant even when the transients are strongly nonexponential as a result of alloy broadening.			
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LUND 1983

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DEEP LEVEL CHARACTERIZATION

The purpose of this experiment is to determine the deep level characteristics of a semiconductor material. This is achieved by measuring the capacitance of a Schottky diode as a function of the reverse bias voltage. The capacitance is measured at various frequencies and the results are plotted on a semi-logarithmic scale. The resulting curves show the presence of deep level traps in the semiconductor material.

The experimental setup consists of a Schottky diode connected to a capacitance meter. The reverse bias voltage is varied from 0V to 10V. The capacitance is measured at frequencies of 100 Hz, 1 kHz, and 10 kHz. The results are plotted on a semi-logarithmic scale of capacitance versus reverse bias voltage.

The results show that the capacitance decreases as the reverse bias voltage increases. This is due to the depletion of the semiconductor material. The presence of deep level traps is indicated by the non-linear nature of the capacitance-voltage curves.



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EPITAXIAL GROWTH AND DEEP LEVEL CHARACTERIZATION OF $\text{GaAs}_{1-x}\text{P}_x$

by

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The subjects covered in this thesis are the epitaxial growth of the ternary semiconductor $\text{GaAs}_{1-x}\text{P}_x$ using metalorganic vapour phase epitaxy (MOVPE), electrical and optical characterization of the grown material, and tailor growth of MOVPE material for systematical studies of deep levels. The results obtained are presented in the following seven papers:

I Electrical and optical properties of deep levels in MOVPE grown GaAs

J. Crystal Growth 55, (1981) 164.

II Alloying mechanisms in MOVPE $\text{GaAs}_{1-x}\text{P}_x$

J. Crystal Growth 61, (1983) 425.

III Organometallic epitaxial growth of $\text{GaAs}_{1-x}\text{P}_x$

J. de Physique 12, (1982) C5-325.

IV Electrical and optical properties of MOVPE grown $\text{GaAs}_{1-x}\text{P}_x$

Accepted for publication in Il Nuovo Cimento (1983).

V Deep level transient spectroscopy evaluation of nonexponential transients in semiconductor alloys

J. Appl. Phys. 54, (1983)

VI Electrical properties of Fe in GaAs

J. Appl. Phys. 54, (1983) 814.

VII The metastable state of EL2 in the $\text{GaAs}_{1-x}\text{P}_x$ alloy system

Manuscript (to be published).

The co-authors are H.G. Grimmeiss in papers I-VII, M. Kleverman and L.-Å. Ledebø paper VI, L. Samuelson papers I-V and VII, and H. Titze papers I, III and IV.

The introduction includes a short background describing the reasons for the interest in the epitaxial growth of III-V semiconductors and the motivation behind the study of deep levels. Some of the properties of $\text{GaAs}_{1-x}\text{P}_x$ which are of interest in this study will also be described. The growth mechanisms and alloying mechanisms of MOVPE $\text{GaAs}_{1-x}\text{P}_x$ are discussed and, after a presentation of the different characterization techniques used in this work, two theoretical models used to describe the properties of defects are introduced. Finally motivation is given for the study of the deep donor level, EL2, in GaAs and $\text{GaAs}_{1-x}\text{P}_x$.

Background

The reason for the explosive development of semiconductor physics and device technology is the ability to grow and, in a controlled way, dope semiconductors. As a result of massive research efforts in this field, it has been possible to construct sophisticated devices like charge coupled devices (CCD) and monolithic integrated circuits of high complexity.

So far, the dominating semiconductor material has been silicon. Although the group IV semiconductor silicon shows many good qualities it lacks others. For instance, the band gap, of about 1.1 eV, excludes Si as a suitable material for visible light emitting diodes (LEDs), which require band gaps larger than about 1.8 eV. Si is also excluded as a suitable material for an injection laser because of the indirect band gap.

This has stimulated interest in III-V semiconductors. Some of these compound semiconductors show excellent electrical and optical properties due to their high electron mobility and direct band gap. The band gaps are quite different for different compounds, from 0.17 eV for InSb up to 2.4 eV for AlP, and some have band gaps suitable for visible LED applications. It is also possible to form ternary and quaternary alloys, such as $\text{GaAs}_{1-x}\text{P}_x$ and $\text{Ga}_{1-y}\text{In}_y\text{As}_{1-x}\text{P}_x$, which allows the size of the band gap to be changed by varying the composition of the alloy. In an LED or an injection laser where the wavelength of the emitted photons is determined by the band gap, this band gap variation can be used to obtain suitable wavelengths for different applications.

For many years, the limiting factor in III-V semiconductor physics was the problem of preparing single crystals of high quality. Good progress has been made in growth technology in the areas of both bulk crystals and epitaxial films. The high quality obtained in epitaxial films is, to a large extent, due to the development of new growth techniques such as liquid phase epitaxy (LPE), molecular beam epitaxy (MBE) and metalorganic vapour phase epitaxy (MOVPE) [1,2]. These epitaxial techniques have already contributed to both semiconductor physics and to device technology by introducing and developing new concepts such as injection lasers, quantum wells and two dimensional electron gases.

MOVPE, probably the most versatile method for producing III-V semiconductors, has been used in the growth of several combinations of group III and group V elements [3,4]. There are, however, still III-V alloy systems where the MOVPE technique has not been thoroughly investigated. Furthermore, since most investigations and reports of MOVPE growth have been for device purposes, the research and understanding of growth mechanisms is in many cases incomplete. This lack of understanding about the growth and, especially, alloying mechanisms of $\text{GaAs}_{1-x}\text{P}_x$ provides important motivation for the studies described in this thesis.

Defects (impurities, vacancies, interstitials etc.) determine many of the important properties of semiconductors [5]. Some defects are desirable, such as shallow dopant impurities and radiative recombination centres, while others are undesirable, for example non-radiative recombination centres in LEDs. Success in epitaxial growth of material of high electrical and optical quality demands a good understanding of the physics of, and the incorporation of, defects. Different defects in a crystal give rise to different electronic energy levels. Some of these energy levels fall in the forbidden band gap. Such energy levels where the ground state of the electronic wavefunction is considerably delocalized, effective mass theory is valid, and the ionization energies are small (compared with the band gap), are called shallow levels. Defects with a more localized ground state of the electronic wavefunction require larger ionization energies, and the energy level is thus located further from the band. This type of defect is called a deep level, and the theoretical treatment is far more complex than for shallow levels. In fact, knowledge about many of these deep levels is, in spite of experimental and theoretical efforts, still incomplete. Characterization of deep levels in $\text{GaAs}_{1-x}\text{P}_x$ is therefore an important part of the work which will lead to a better understanding of

defects in III-V semiconductors.

One approach towards obtaining more information on deep levels is to study the behaviour of the centre as a function of alloy composition. MOVPE is probably the most suitable technique for the tailor growth of material for systematic studies of deep levels in III-V semiconductors. This provides further important motivation for the work described in this thesis.

The $\text{GaAs}_{1-x}\text{P}_x$ material

Both GaAs and GaP crystallize in the cubic zinc blende structure shown in Fig. 1. The length of the unit cube is 5.45 Å in GaP and 5.65 Å in GaAs ($T=300$ K). For these face-centred lattices the first Brillouin zone (in reciprocal space) is shown in Fig. 2, and the most important paths, when describing e.g. band structures, are those drawn from the zone centre Γ to the high symmetry points X, L etc. (see Fig. 2).

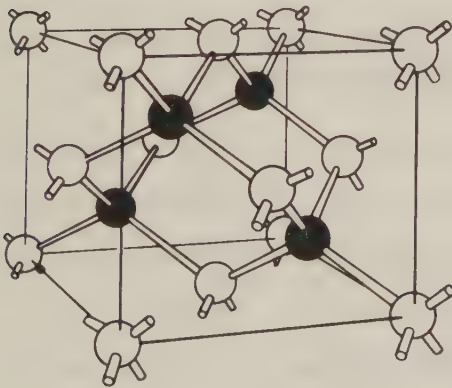


Fig. 1: Conventional cubic cell for the zinc blende lattice. This is the structure of both GaAs and GaP.

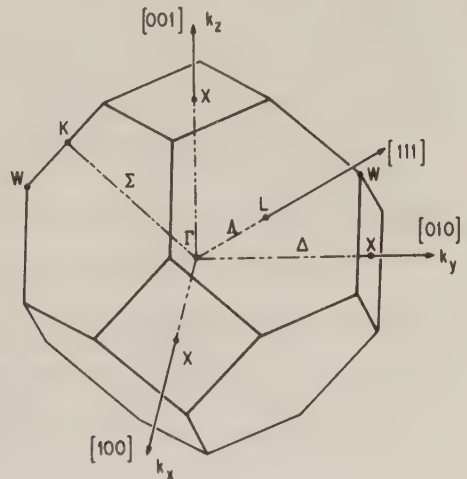


Fig. 2: The first Brillouin zone for the zinc blende lattice. Important symmetry points are indicated.

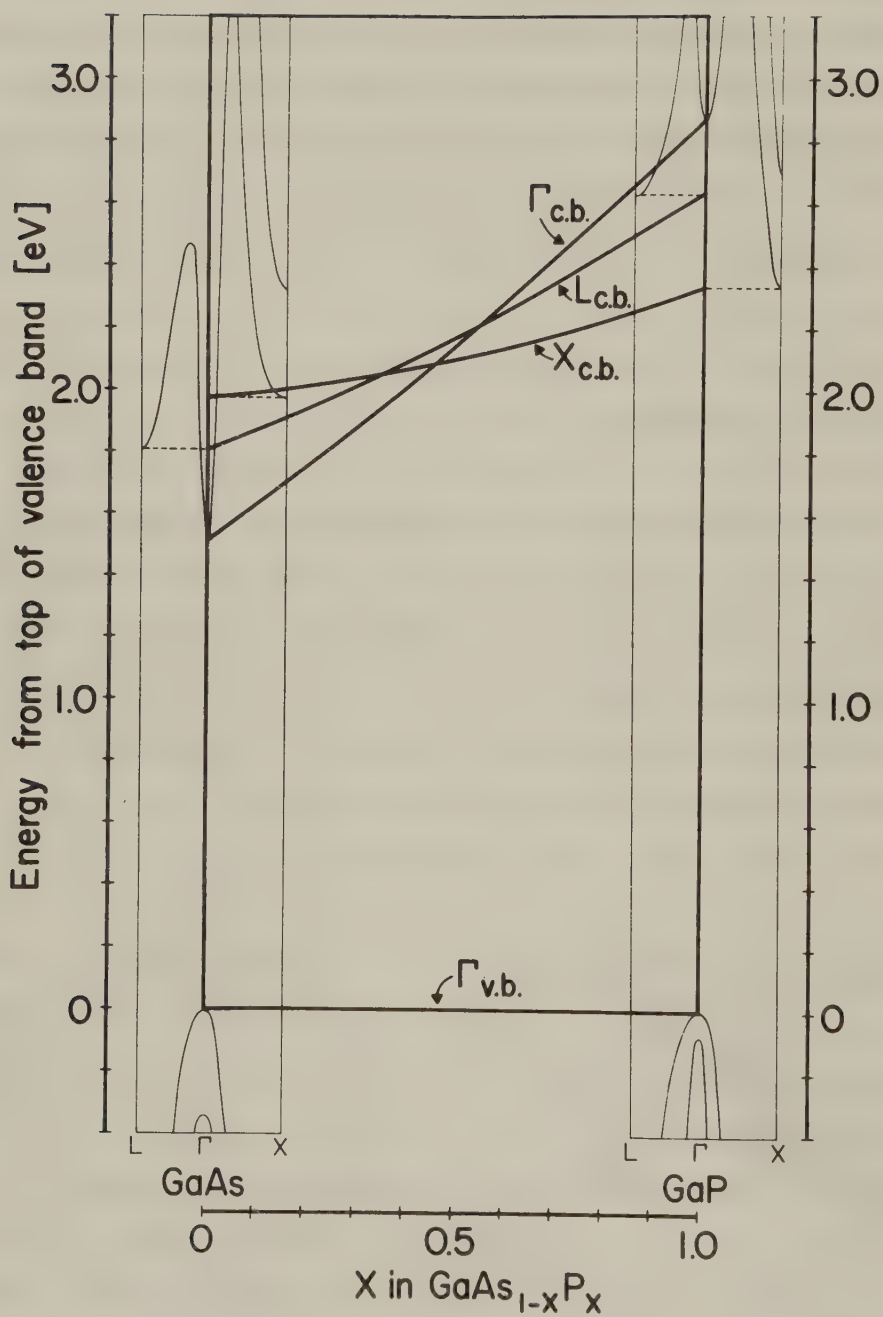


Fig. 3: The electronic band structures ($T=77\text{ K}$) for GaAs and GaP (thin lines). The variations of the band extrema with alloying are also indicated (thick lines).

The band structures of GaAs [6] and GaP [7] are shown in Fig. 3. The smooth variation of the Γ , L and X conduction band minima with varying alloy composition is also illustrated in this figure [8]. Since GaAs is a direct band gap material and GaP an indirect band gap material, the optical and electrical properties are quite different. For the part of the $\text{GaAs}_{1-x}\text{P}_x$ alloy system with a direct band gap ($x < 0.46$) the probability for optical band-band processes is large. The corresponding processes in the indirect alloy system involve momentum conserving phonons, and are thus less effective. A similar trend is shown in the electrical properties. The high mobility observed in GaAs decreases slowly towards the band cross-over, where an abrupt decrease is observed [9].

MOVPE growth of $\text{GaAs}_{1-x}\text{P}_x$ —

Since the first demonstration of MOVPE growth of GaAs [10] the technique has proven to be suitable for the growth of most III-V binary and multinary compounds [2-4]. The inherent advantages of the technique are: a simple system with one heated zone, the ability to easily control the vapour-solid distribution, versatility and the possibility of large-scale production. The most severe disadvantages are: carbon contamination, other impurities in the starting materials, the adduct formation between In alkyls and phosphine and arsine, and the use of poisonous gases.

The equipment for MOVPE growth used in this work is shown schematically in Fig. 4 [11]. This versatile apparatus has sources for the group III atoms Ga, In and Al, and the group V atoms As and P. S and Zn are used as dopants for n- and p-type doping respectively. The group III metalorganic sources (such as $(\text{CH}_3)_3\text{Ga}$ = TMG) are kept liquid in temperature-controlled baths and hydrogen, as a carrier gas, is bubbled

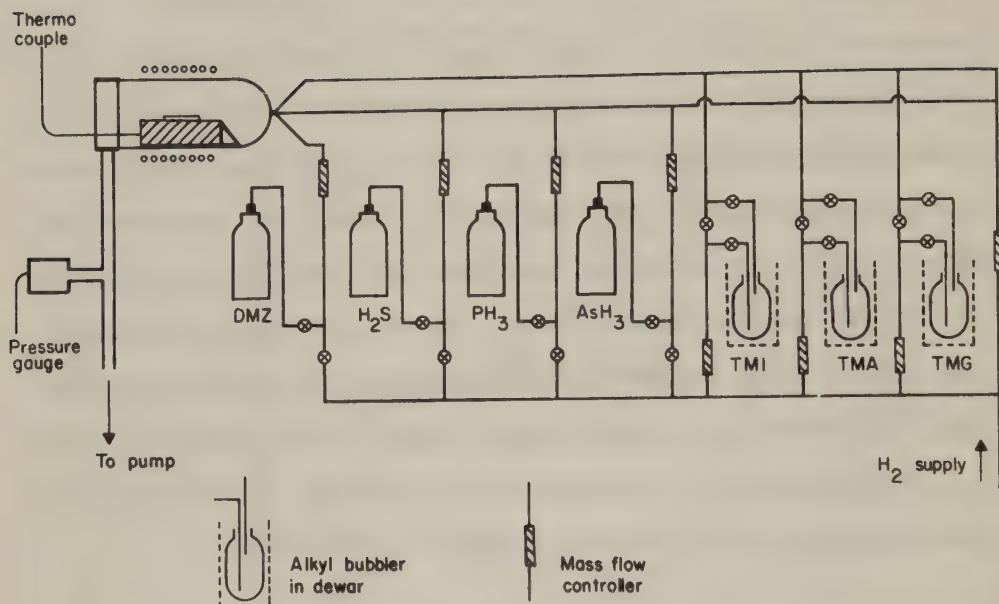


Fig. 4: Schematic diagram of growth apparatus.

through the liquid. The reactive gases (e.g. TMG, AsH_3 and PH_3) are mixed just prior to the entrance of the quartz cell where the substrate is in contact with an RF-heated graphite susceptor.

The growth of GaAs can be described using a simple model [12,13] based on the observed temperature independence of the growth rate, which suggests a diffusion limited growth process. At growth temperature (600-800 °C) the injected TMG molecules are completely pyrolyzed in a homogeneous gas-phase reaction, while the AsH_3 molecules are mainly decomposed by a catalyzed reaction at the surface. The free Ga atoms diffuse through an (assumed) stagnant boundary layer to the surface region where GaAs is formed. Assuming that Fick's law is applicable for the Ga flux through the boundary layer, one finds that the growth rate is proportional to the TMG partial pressure, as is observed experimentally.

The details of the formation of GaP have not been so well investiga-

ted [10,14-16], but a growth mechanism similar to that of GaAs is assumed. This is in fact supported by the reported linear dependence of the growth rate on TMG partial pressure and the temperature independence of the growth rate. Furthermore, since phosphides are thermodynamically more stable than arsenides [13], it is not surprising to find that the growth of high quality GaP requires higher temperatures than for GaAs.

For the formation of $\text{GaAs}_{1-x}\text{P}_x$ alloys [17,18], the growth mechanisms are assumed to be essentially the same as for the binary constituents. A new problem which arises is the vapour-solid distribution of the group V elements As and P. As shown in Fig. 5, the relation between the composition in the vapour phase and in the solid phase is found to be temperature dependent. By using a simple model for the incorporation of competing atoms, it is possible to fit the experimental data with a theoretical expression (papers II and III)

$$\frac{[\text{As}]_s}{[\text{As}]_s + [\text{P}]_s} = (1 + (\beta[\text{P}]_v)/(\alpha[\text{As}]_v))^{-1} \quad (1)$$

where $[\text{As}]_{s,v}$ and $[\text{P}]_{s,v}$ refer to the concentrations of As and P in the

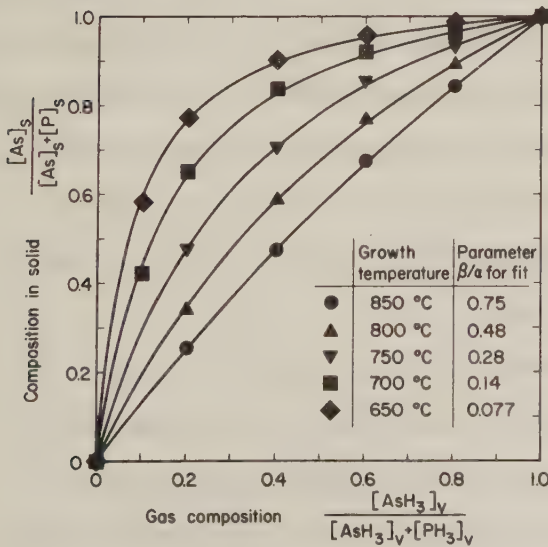


Fig. 5: Experimental data on the composition of the solid plotted vs that for the vapour. The experimental points have been fitted with the theoretical expression given by Eq. 1 for each growth temperature.

solid and vapour phases, and α and β contain the decomposition rates, the recombination rates in the vapour or at the surface and the desorption of atoms adsorbed at the surface, for As and P respectively.

Characterization techniques

For evaluation of the growth process and crystal quality [19], a scanning electron microscope (SEM) has been a standard tool for measuring growth rates, studying interfaces and determining surface quality. In the EBIC (electron beam induced current) mode the position and the quality of the p-n junctions have been studied. The alloy composition and the concentration gradient in the graded layer were determined by electron beam induced x-ray analysis in the SEM. A new technique, based on this microprobe analysis, to determine alloying mechanisms in crystal growth is presented in this thesis (paper II).

In order to characterize the material properties [19], optical characterization methods such as absorption, injection-luminescence and photo-luminescence were used. Information on the optical quality, for example the band edge emission and band gap properties, could thus be obtained. The free carrier concentration of the materials was determined by $1/C^2$ -V measurements and Hall effect measurements. The latter technique also gives information on the mobility.

Defects with radiative recombination were studied with low temperature photo-luminescence techniques.

For the deep level characterization various forms of junction space charge techniques [19-21] were used, such as dark capacitance measurements, deep level transient spectroscopy (DLTS) [22] and photocapacitance measurements including both constant voltage and constant capaci-

tance techniques. By using different forms of junction techniques it is often possible to obtain detailed information on the excitation and de-excitation processes via a specific defect. DLTS is a powerful method not only for rapid surveys of processed material but also for scientific investigations. The method has been questioned as a tool for deep level studies in alloys because of the non-exponential behaviour of the thermal transients. In this thesis (paper V) an evaluation technique is presented which extends the DLTS method to include deep levels in alloys.

Models for defects

For defects with very delocalized wavefunctions, as is the case for shallow impurity levels, a theoretical model, the effective mass theory (EMT) [23], is available. In this model the problem is simplified to the case of an electron bound to a Coulomb potential. Also for the opposite situation with very localized wavefunctions, the crystal field theory (CFT) [5,24] is available. In the CFT it is assumed that the defect in the crystal is only affected by the symmetry of the host via the electrostatic potential from the nearest neighbours. The simple pictures of these extreme cases, the hydrogen atom model for defects with very delocalized wavefunctions and the molecular model for defects with very localized wavefunctions, are illustrated in Fig. 6.

For most defects, neither of these models is applicable, and the problem of modelling deep levels must be treated by considering both the crystal properties and the defect properties at the same time. A review of different methods for such deep level calculations is given in Ref. 5.

In the effective mass theory a many body problem (the defect and the crystal) is reduced to the problem of one particle (the defect) in a

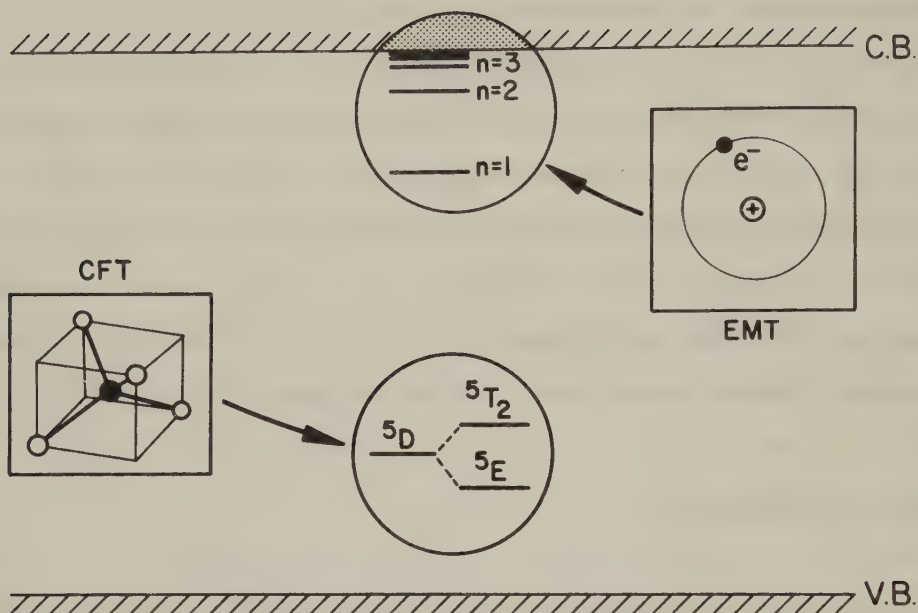


Fig. 6: An illustration of the models discussed in the text.

dielectric medium. The Schrödinger equation for this reduced problem is the same as for a hydrogen atom, scaled by the dielectric constant ϵ and the relevant effective mass m^* . In this, the most simple form of the EMT, the energy values below the ionization limit are given by

$$E_n = \frac{1}{n^2} \cdot \frac{m_0}{m^* \epsilon} \cdot 13.6 \text{ (eV)} \quad (2)$$

By improving the theory to include crystal anisotropy [25], inter-valley mixing in indirect semiconductors [25] and effects of degenerate band edges [26], the EMT has been successful in predicting the positions of the excited states of shallow donors and acceptors with great precision [27]. The EMT has also proven to be successful in the calculation of the excited states of the deep chalcogen levels in Si [28].

As an example of the crystal field theory, and as a complement to paper VI in this thesis, a short summary of the CFT, applied to Fe in GaAs

[29], will be given.

When iron substitutionally occupies a Ga site in GaAs it forms an acceptor. The transition element Fe, with outer shell configuration $3d^6 4s^2$, completes the crystal bonding and leaves an $Fe^{3+}d^5$ defect in the lattice. This defect can only be observed when it has accepted an electron, i.e. in the $Fe^{2+}d^6$ state. The lattice interaction on the defect is, in the CFT, replaced by electrostatic point charges on the four nearest neighbour sites. This field simulates the crystal potential on the $3d^6$ electrons in the defect, and lowers the full rotation symmetry of the free atom to T_d (tetrahedral) symmetry. The total Hamiltonian can now be expressed as $H = H_0 + H_{cr}$ where H_0 is the free "d⁶-atom" energy operator in the many-electron central field approximation and H_{cr} is the crystal field operator which acts as a perturbation. The energy eigenvalue of H_0 on a "d⁶-atom" has already been calculated by atomic physicists, and the ground state is the 5D term. This level is, according to group theory, split in a tetrahedral symmetry to 5E and 5T_2 (5E is lower in energy). The splitting, Δ , can be calculated by using H_{cr} in a perturbation calculation. The absolute values obtained are very uncertain because of the crude approximation, and Δ is often considered as an experimentally determined parameter. By taking more terms into consideration in the calculation, for example spin-orbit interaction (LS), further splitting can be found. (The $H_0 > H_{cr} > H_{LS}$ situation is often referred to as the intermediate crystal field situation.) The CFT is only applicable to the internal splitting of the term values. Calculating the absolute energy positions of the terms is a much more complicated problem which requires that the crystal interaction on the defect be treated in detail [5].

The EL2-level

The most important and most investigated defect in GaAs is probably the mid-gap deep donor level EL2, which has been found to be the dominant electron trap in both bulk grown material and VPE material (including the MOVPE material in this work) [30,31]. The level has also been labelled "0"-level since it was originally attributed to oxygen.

The importance of the EL2-level from the device point of view, is the compensating role it plays in undoped liquid-encapsulated Czochralski (LEC) and Bridgman- grown bulk material, leading to the formation of semi-insulating (SI) GaAs [32]. Such material is promising for further processing, such as ion implantation into the SI material for the fabrication of GaAs integrated circuits.

The EL2-level shows very exciting physical properties. For instance, when a photocapacitance experiment is carried out on a normal deep donor level, the signal increases monotonically to a final value. Performing the same experiment on the EL2-level at a low temperature ($T=77$ K) the signal increases, but instead of reaching the final value it decreases again to the initial value. Once transformed to this state, the EL2 centre is inaccessible to further extrinsic illumination. This behaviour indicates the existence of a metastable state of the same charge as the ground state [33,34]. In spite of the great effort put into the understanding of the EL2-level in the last decade, neither the microscopic nature of the defect nor an empirical model which can explain all experimental data is available.

In this thesis two different approaches to obtaining more information on the microscopic structure of the EL2-level have been used. Both of these approaches require specially grown materials, for which the MOVPE

technique is especially suitable. In the first approach, the formation of the EL2-level was studied by systematical variations of the thermodynamical conditions at growth, which gave information on the native defect incorporation in the material (paper I). The second approach was the growth of $\text{GaAs}_{1-x}\text{P}_x$ crystals under suitable growth conditions to form the corresponding deep level in the alloys (paper IV, V and VII). The purpose was to study the different properties of the defect as a function of alloy composition, and in the extension of this, to study the defect in GaP where its chemical nature may already be known.

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THE METASTABLE STATE OF EL2 IN THE $\text{GaAs}_{1-x}\text{P}_x$ ALLOY SYSTEM.

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Abstract

The photocapacitance quenching effect of EL2 previously observed in GaAs has been studied in the $\text{GaAs}_{1-x}\text{P}_x$ alloy system and optical and thermal properties of the metastable state have been determined. It is shown that the spectral distribution of the optical transition rate for the population of the metastable state ($\text{EL2}^0 \rightarrow \text{EL2}^*$) is nearly independent of the alloy composition. A previously unknown temperature dependence of the spectral distribution of the transition $\text{EL2}^0 \rightarrow \text{EL2}^*$ in GaAs is reported and a possible explanation is discussed.

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I. INTRODUCTION

Owing to its technical importance and interesting physical properties the EL2 center in GaAs has been the subject of many detailed studies during the last decade [1]. Since both origin and properties of the center are poorly understood a new approach for a better understanding of this center is attempted in this paper by studying properties of the EL2-level as a function of the composition in the alloy system $\text{GaAs}_{1-x}\text{P}_x$ [2,3]. One of the most intriguing properties of GaAs:EL2 is the existence of a metastable state [1,4,5]. In samples with low carrier concentrations this state can be populated, at sufficiently low temperatures ($T \lesssim 140$ K), by illuminating the sample with photons in the energy range $1.0 \lesssim h\nu \lesssim 1.3$ eV. Once populated the metastable state is inaccessible to extrinsic light, and can only be depopulated thermally by heating the sample to temperatures above about 110 K. Another possibility to depopulate the state is to let free carriers interact with the metastable state. This process is active already for temperatures below 110 K.

The purpose of this paper is to present a study of the metastable state of EL2 in the $\text{GaAs}_{1-x}\text{P}_x$ alloy system. Photocapacitance data are presented showing how the optical and thermal properties of the metastable state of EL2 change with composition. It will also be shown that the depopula-

tion process of the metastable state can be expressed in one formula, valid for the part of the alloy system investigated. Furthermore, data are presented which reveal for the first time an unexpected temperature dependence of the spectral distribution of the optical cross section for the population of the metastable state.

II. EXPERIMENTAL

All measurements have been performed on undoped n-type $\text{GaAs}_{1-x}\text{P}_x$ epitaxial layers grown on GaAs substrates by Metal-Organic Vapour Phase Epitaxy (MOVPE). Details about the epitaxial growth and the electrical and optical properties of the layers have been published previously [2,6].

The electrical characterization of the samples included capacitance-voltage (C^{-2} -V) measurements for the determination of carrier concentrations ($|N_d - N_a|$) and DLTS (Deep Level Transient Spectroscopy) [7,8] measurements to evaluate deep level concentrations (N_T). Photocapacitance measurements were performed in a temperature controlled cryostat (10-300 K) using a Boonton 72BD capacitance meter and a Zeiss MM3 prism monochromator. A Bauch and Lomb grating monochromator was employed for broad band illumination.

Some important parameters of the samples investigated are collected in Table I.

III. EXPERIMENTAL RESULTS

A. Background

Even though photocapacitance kinetic of the EL2 center has been described previously [1,5,9] it may be useful for the understanding of this paper to give a brief summary of the results found in GaAs. In an n-Schottky

diode at temperatures higher than $T \approx 150$ K but lower than ≈ 250 K (in order to avoid thermal emission processes) the EL2 centers respond to optical excitation as a single deep midgap level at energy position $E_C - E_T \approx 0.75$ eV. Choosing the initial condition in a reversed biased diode such that the EL2 levels are occupied with electrons, and illuminating the diode with photons of energy $h\nu$ (where $E_C - E_T \leq h\nu < E_C - E_V$), the observed change in capacitance is given by [10] (notations according to Ref. [10]).

$$\Delta C(t) = \frac{e_n^0}{e_n^0 + e_p^0} N_{TT} \cdot \frac{1}{2} \left(\frac{\epsilon \epsilon_0 \cdot A^2 \cdot q}{2(V_D - V_R)N_d} \right)^{\frac{1}{2}} [1 - \exp(-(e_n^0 + e_p^0) \cdot t)] \quad (1)$$

It is readily seen that the time dependence of the capacitance signal is exponential with the time constant $\tau_1^{-1} = e_n^0 + e_p^0$. This behaviour corresponds to curve (a) in Fig. 1. Performing the same experiment at temperatures lower than 100 K, the time dependence of the photocapacitance signal becomes more complicated (curve (b) in Fig. 1). Before reaching the steady-state occupancy observed at $T \geq 150$ K ($n_T(\infty) = (e_p^0/(e_n^0 + e_p^0)) N_T$) the capacitance signal decreases with a time constant τ_2 approaching the initial capacitance value. This is the photocapacitance quenching effect of EL2 in GaAs. From several arguments [9] it has been concluded that the effect is caused by the transformation of the "normal" EL2 center ($EL2^0$) into a metastable state ($EL2^*$) of the same charge state. Once populated the state is no longer affected by extrinsic light. The spectral distribution of the optical transition rate for the population of the metastable state is shown in the insert of Fig. 1. The regeneration (from the metastable state, $EL2^*$, to the normal state, $EL2^0$) is often described by an empirical regeneration rate r , which in turn can be divided into the thermal regeneration rate,

$$r^{th} = r_0^{th} \exp(-\Delta E^{th}/kT) \quad (2)$$

and the electron induced regeneration rate

$$r^{el} = \sigma^{el} \cdot n \cdot \langle v_{th} \rangle = \sigma_0^{el} \cdot n \cdot \langle v_{th} \rangle \exp(-\Delta E^{el}/kT) \quad (3)$$

($\langle v_{th} \rangle$ is the thermal velocity of free electrons, and σ^{el} is the capture cross section)[5,9]. Both rates are temperature activated ($\Delta E^{th} \approx 0.34$ eV and $\Delta E_a^{el} \approx 0.11$ eV), and in addition, r^{el} is proportional to the free electron concentration n .

B. Identification of EL2 in $\text{GaAs}_{1-x}\text{P}_x$

The identification of the deep donor observed in $\text{GaAs}_{1-x}\text{P}_x$ as being the EL2 center observed in GaAs is based on three arguments. First, thermal emission rates studied as a function of temperature show continuous trends with alloy composition, both for activation energies and thermal emission rates [3]. Second, we observe continuous variations of details in the photoionization cross sections when the alloy composition is varied. Third, broad band illumination ($h\nu \approx 1.15$ eV) at low temperatures causes the characteristic photocapacitance quenching effect of the studied center for $x \leq 0.26$ as for EL2 in GaAs (Fig. 2). The quenching effect was not observed for $x \geq 0.38$, although the EL2 center seems to be still present according to the first two arguments.

C. Spectral distribution

Fig. 3 shows the spectral efficiency $(\phi \cdot \tau_2)^{-1}$ vs $h\nu$ of the photocapacitance quenching effect for different alloy compositions. Low effective capture cross sections for the $\text{EL2}^0 \rightarrow \text{EL2}^*$ transition made it impossible to obtain reasonable resolution and to monitor simultaneously the full transient. τ_2 was therefore deduced (as in Ref. [9] and Ref. [11]) by the initial slope of the quenching transient i.e.

$$\tau_2^{-1} = \frac{1}{\Delta C_0} \cdot \left. \frac{d\Delta C}{dt} \right|_{t=0} \quad (4)$$

For notation see the insert of Fig. 3. Each spectrum has been normalized at its peak position.

D. Thermal regeneration

As mentioned earlier the thermal regeneration rate of the metastable state of GaAs:EL2 ($EL2^* \rightarrow EL2^0$) is temperature dependent [5,9] and can be expressed as shown in Eq. 2. r^{th} has been measured using the isothermal method [9] illustrated in Fig. 1. After having transformed all $EL2^0$ states into $EL2^*$ states r^{th} has been obtained in GaAs by measuring the difference $\Delta C(t_d = \infty) - \Delta C(t_d)$ (which is proportional to $[EL2^*]$, see Fig. 1) as a function of t_d , the time after the light source has been removed (see Fig. 4). E^{th} can then be calculated from an Arrhenius plot of r^{th} . In contrast to GaAs a non-exponential time dependence is observed in $GaAs_{1-x}P_x$ (see Fig. 4). An apparent regeneration rate $r^{th-} = r_0^{th-} \exp(\Delta E^{th-}/kT)$, (which becomes the true regeneration rate r^{th} for $x=0$) has therefore been used in alloys by taking the time t_d^- at which the capacitance signal decreased to $\Delta C(t_d = \infty) - \Delta C(t_d^-) = \Delta C(t_d = \infty)/e$ and defining $r^{th-} = 1/t_d^-$ (see Fig. 4). It is interesting to note that the Arrhenius plots of r^{th-} for all compositions investigated gave straight lines (Fig. 5) from which the thermal activation energy ΔE^{th-} readily could be deduced. The activation energy ΔE^{th-} was found to be independent of the definition of r^{th-} , i.e. $\Delta E^{th-} = \Delta E^{th}$ is a well defined activation energy. The values obtained for r_0^{th-} and ΔE^{th} in samples of different compositions are summarized in Table II.

E. Electron induced regeneration

In GaAs the regeneration of $EL2^*$ is accelerated by the presence of free electrons. Similar effects have been observed in our alloys for $x \leq 0.08$. For larger values of x the measurements were disturbed by the presence of another deep center which was thermally active in the temperature

range in which the measurements were performed. In principle the electron induced regeneration rate (Eqn. 5) can be measured by a similar method as the one described in Section III D. However, due to experimental difficulties a simpler measuring technique has been used. Keeping the electrical pulse length t_e constant, $\Delta C(t_e = \infty) - \Delta C(t_e)$ has been measured as a function of temperature. Assuming that $[EL2^*](t_e) = [EL2^*](t_e=0) \cdot \exp(-n \langle v_{th} \rangle \sigma^{el} t_e)$ and $\sigma^{el} = \sigma_0^{el} \exp(-\Delta E^{el}/kT)$ it is easy to see that a plot of $\log(\log([EL2^*](t_e=0)/[EL2^*](t_e)))$ vs. $1/kT$ yields the activation energy ΔE^{el} . The results obtained are summarized in Table II.

IV. DISCUSSION

The spectral distribution for the optical transition from the normal to the metastable EL2 state has been measured in previous studies by three different methods. The first method implied phot capacitance measurements (80 K) in epitaxial layers [9] (as in this work), the second method used the fatigue effect of the 0.63 eV photoluminescence (4 K) in semiinsulating bulk GaAs [11] and in the third method the difference between absorption and photocurrent spectra (80 K) in n-type bulk GaAs was studied [12]. The different methods gave similar results, but small differences are observed. Our peak position at 100 K was about 30 meV lower in energy than the phot capacitance data at 80 K [9], which in turn were about 30 meV lower than the 4 K photoluminescence data [11]. To investigate whether these discrepancies reflect a temperature dependence or whether they are caused by differences in experimental conditions, the spectra of the $EL2^0 \rightarrow EL2^*$ optical cross section were measured at different temperatures (Fig. 6a). For comparison the 4 K photoluminescence data are also plotted, however, with an adjusted peak value. The data clearly reveal a shift in peak position and a change of the onset of the low energy side. This is even more clearly seen in Fig. 6b where the

4 K photoluminescence data have been subtracted from the photocapacitance measurements at higher temperatures. This temperature behaviour may suggest a splitting of either the initial state or the final state, i.e. either a difference in absorption strengths with temperature or a difference in branching efficiency from an intermediate excited state to the metastable state, respectively. In fact, the existence of an intermediate excited state has previously been proposed [12].

If the temperature dependence in Fig. 6 is caused by a splitting in the initial state (i.e. the $EL2^0$ ground state) the same temperature dependence should be observed in absorption experiments. However, neither a shift to lower energies nor an over-all increase in the absorption coefficient with increasing temperature has been reported [12]. Judging from this comparison with absorption data the temperature dependence in Fig. 6 may indicate the presence of a more complex intermediate state (states) with a temperature dependent transfer to the metastable state.

The shift in the optical cross section spectrum with temperature agrees well with the data in Ref. [9] and Ref. [11], but not with those in Ref. [12] where the peak position is slightly higher in energy ($h\nu=1.18$ eV, $T=80$ K). Furthermore, it is claimed in Ref. [12] that the peak position shifts to higher energies with increasing temperatures. These discrepancies may be due to uncertainties in subtracting results from two different measuring techniques.

Defects with similar electronic properties as EL2 in GaAs have been observed by dark capacitance methods in GaInAs [13], AlGaAs [14-16] and in GaInAsP [17]. To the best of our knowledge, no EL2 photocapacitance quenching effect in any alloy or any material other than GaAs has been reported so far. Since the center behaves similarly in GaAs and in $GaAs_{1-x}P_x$ (for $x \lesssim 0.3$) it

seems most likely that the effect does exist also in other alloys, at least for small x values.

The similarity of the optical cross section, the thermal regeneration rate and the electron induced regeneration rate of EL2 in $\text{GaAs}_{1-x}\text{P}_x$ alloys and in GaAs suggests that the microscopic nature of the defect is similar.

Assuming the existence of an intermediate state for the $\text{EL2}^0 \rightarrow \text{EL2}^*$ transition the absorption coefficient for the intercenter transition given in Ref. [12] corresponds then to the absorption between EL2^0 and the intermediate state. This absorption coefficient and the one for the ionization process are of the same magnitude in GaAs ($\approx 1 \text{ cm}^{-1}$ [12]).

The ratio between the optical cross section for ionization and the optical cross section for the $\text{EL2}^0 \rightarrow \text{EL2}^*$ transition, is estimated from our phot capacitance measurements to be ≈ 10 (the ratio between τ_1 and τ_2 in Fig. 1). As already noted in Ref. [12] this observation suggests that the excited state acts as an intermediate branching state for the $\text{EL2}^0 \rightarrow \text{EL2}^*$ transition. Furthermore, since the ratio of the probability, for optical ionization of EL2^0 and population of EL2^* (τ_1 and τ_2 in Fig. 2) in $\text{GaAs}_{1-x}\text{P}_x$ is similar to the one observed in GaAs, it is reasonable to assume that the intermediate state acts also in $\text{GaAs}_{1-x}\text{P}_x$ as a branching state for the $\text{EL2}^0 \rightarrow \text{EL2}^*$ transition.

The non-exponential behaviour of the thermal regeneration rate measurements (Fig. 4) is interpreted as an alloying effect, i.e. different EL2 centers have different local surroundings which results in a distribution of time constants. The activation energy, which does not depend on the definition of r^{th} (see Sect. III D), decreases with increasing phosphorous concentration in the alloy (see Fig. 7). This decrease is also observed for the electron induced activation energy ΔE^{el} . Our

data can be summarized in a unifying expression for the thermal and electron induced regeneration rates valid for the investigated part of the alloy system (see Fig. 8);

$$r(x,T) = 2 \cdot 10^{-5} \exp \left[\frac{\Delta E^{th}(x)}{k_B} \left(\frac{1}{T} - \frac{1}{T^{th}} \right) \right] + 9 \cdot 10^{-21} \cdot n \cdot \langle v_{th} \rangle \exp \left[\frac{\Delta E^{el}(x)}{k_B} \left(\frac{1}{T} - \frac{1}{T^{el}} \right) \right] \quad (5)$$

where $T^{th} = 104$ K (from Fig. 8a), $T^{el} = 82$ K (from Fig. 8b), $\langle v_{th} \rangle$ the thermal velocity in cm/s and n is the free carrier concentration in the conduction band (cm^{-3}). The reason for the disappearance of the photocapacitance effect for $x \approx 0.3$ may be due to the lowering of the activation energy ΔE^{th} . Changes in the energy position of the $EL2^*$ level relative to the $EL2^0$ level may make the metastable state energetically unfavourable. Alternatively, increasing (decreasing) the $P(As)$ concentration may reduce the probability of producing the microscopic aggregate responsible for the metastable properties.

V. CONCLUSION

The photocapacitance quenching effect, typical for $EL2$ in $GaAs$, is observed for the corresponding defect in the $GaAs_{1-x}P_x$ alloy system. The spectral distribution of the optical transition rate for the population of the metastable state has been studied for different values of x , yielding remarkably constant spectral shapes. In $GaAs$ the spectrum has been found to be temperature dependent. This temperature dependence has been discussed in terms of splittings in either the ground state or an excited state.

The thermal regeneration rate ($EL2^* \rightarrow EL2^0$) is thermally activated with an activation energy decreasing from 0.36 eV ($GaAs$) to 0.16 eV ($x=0.20$).

The electron induced regeneration rate shows a similar reduction in activation energy. It is also shown that the regeneration rate (thermal and electrical) can be expressed in one single formula valid for the investigated part of the alloy system.

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Table I. Characteristics of the n-type Schottky samples used in this work.

x in GaAs _{1-x} P _x	$ N_d - N_a $ [10 ¹⁶ ·cm ⁻³]	EL2 konc. [10 ¹⁴ ·cm ⁻³]	$\vec{E}$ [10 ⁴ ·V/cm]
0	0.66	2.0	4.5
0.04	0.54	1.9	4.1
0.08	0.62	1.4	4.4
0.20	1.3	0.6	6.5
0.26	1.3	0.7	6.5

Table II. Collected parameters for thermal and electron induced regeneration (EL2* → EL2⁰).

GaAs _{1-x} P _x :EL2		r_0^{th} [s ⁻¹]	ΔE^{th} [eV]	σ^{el} [cm ²]	ΔE^{el} [eV]
x=0	Ref. 9	$2 \cdot 10^{11}$	0.30	$1.4 \cdot 10^{-13}$	0.108
	Ref. 5	$8.6 \cdot 10^{11}$	0.34	$1.9 \cdot 10^{-14}$	0.107
	This work	$3.6 \cdot 10^{12}$	0.36	$2.8 \cdot 10^{-14}$	0.106
x=0.04		$5.1 \cdot 10^7$	0.25	$2.5 \cdot 10^{-17}$	0.060
x=0.08		$2.3 \cdot 10^5$	0.21	$3.3 \cdot 10^{-18}$	0.043
x=0.20		$8 \cdot 10^2$	0.16	-	-

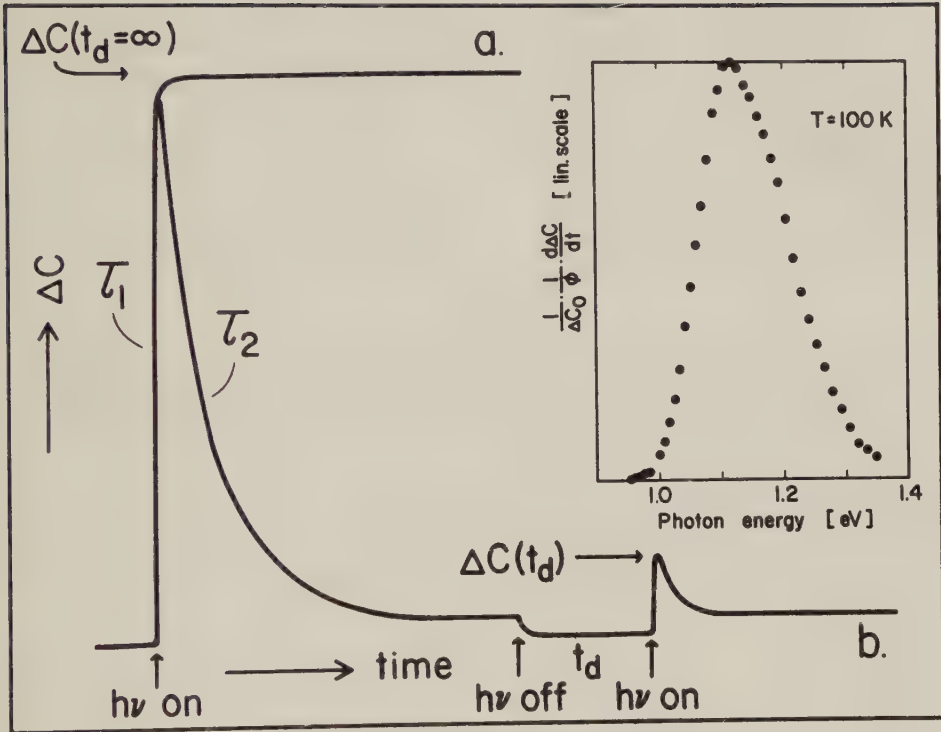


Fig. 1 The photocapacitance signal due to EL2 in GaAs for two different temperatures. Curve (a) corresponds to Eq. 1 ($T=200$ K) and curve (b) shows the photocapacitance quenching effect ($T=77$ K). The spectral distribution for the $EL2^0 \rightarrow EL2^*$ transition at $T=100$ K is inserted. Also illustrated by curve (b) is the measurement of the thermal regeneration as discussed in Sec. III D. $\Delta C(t_d=0)$ is proportional to N_t and $\Delta C(t_d)$ is proportional to the concentration of $EL2^0$ ($[EL2^0]$).

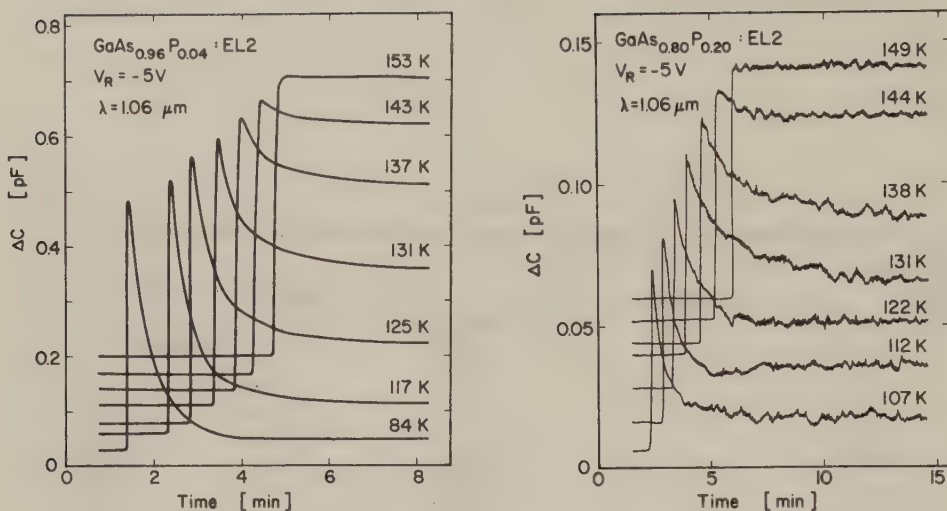


Fig. 2 The photocapacitance quenching effect, typical for EL2 in GaAs, as observed for the corresponding defect for two $\text{GaAs}_{1-x}\text{P}_x$ alloy compositions.

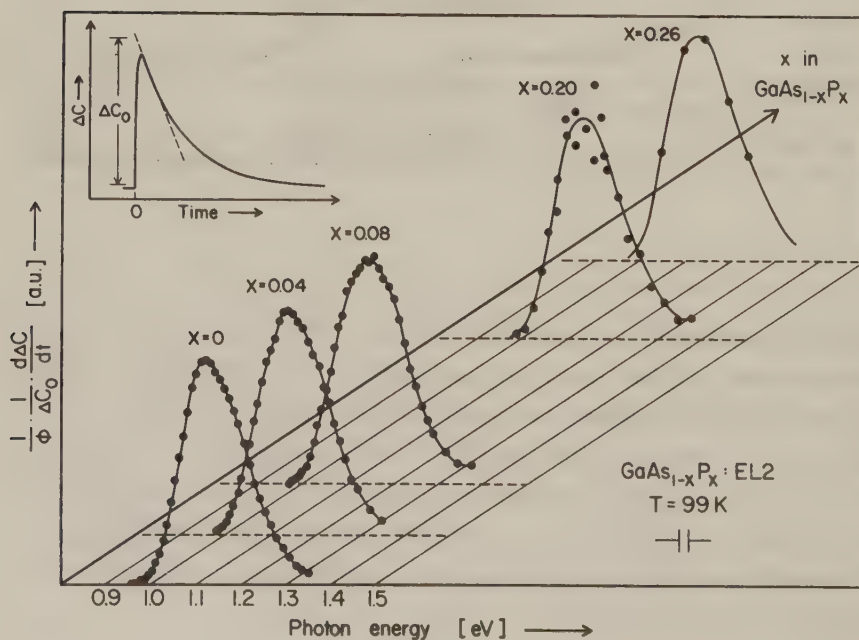


Fig. 3 The spectral distribution of the $\text{EL2}^0 \rightarrow \text{EL2}^*$ optical cross section for different alloy compositions.

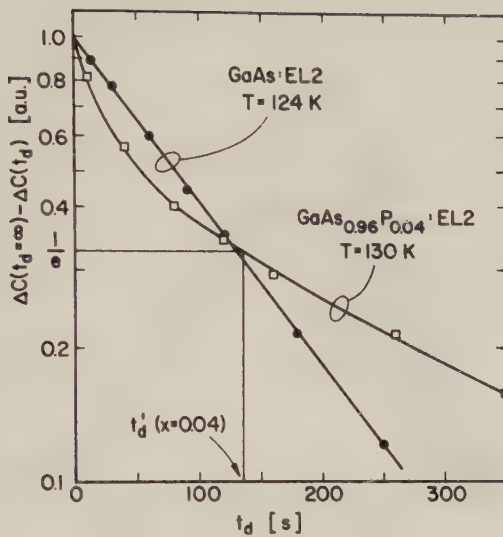


Fig. 4 A plot of the concentration of EL2-centres remaining in the metastable state after waiting different times t_d in the dark. The concentration $[EL2^*]$ is obtained as $\Delta C(t_d = \infty) - \Delta C(t_d)$ where ΔC is the peak-height in the capacitance transient in Fig. 1. A non-exponential behaviour is observed in the alloy. Also indicated is the evaluation of r^{th} in the alloys (see Sec. III D).

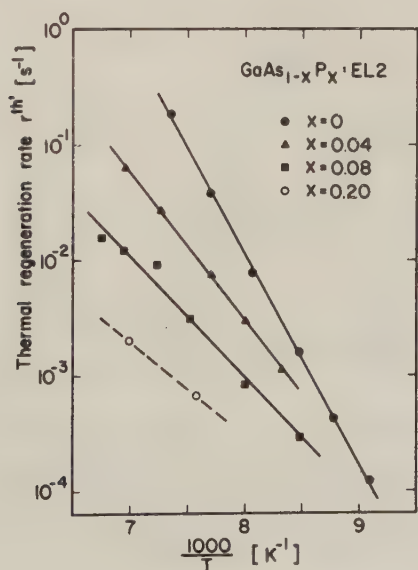


Fig. 5 The logarithm of the thermal regeneration rates, r^{th} , plotted vs. $1/T$ for different alloy compositions.

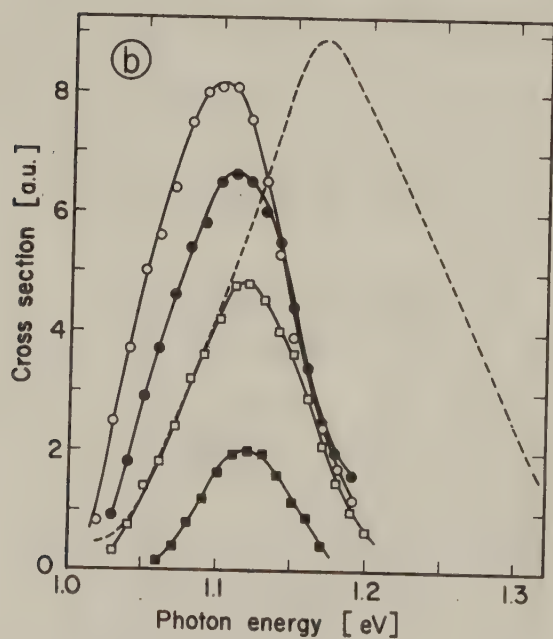
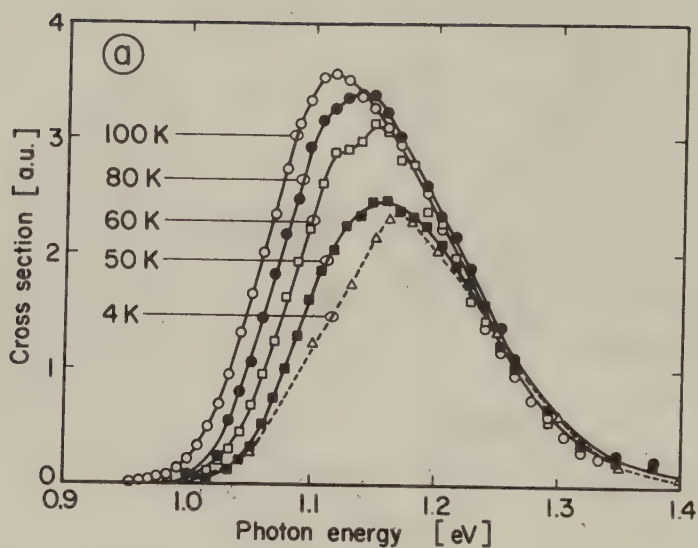


Fig. 6 a) The optical transition rate for population of the metastable state plotted vs photon energy for different temperatures (full lines). The dashed line is a low temperature spectrum (4 K) obtained from photoluminescence quenching [11].

b) Same data as in a) but with the 4 K-spectrum subtracted from each spectrum.

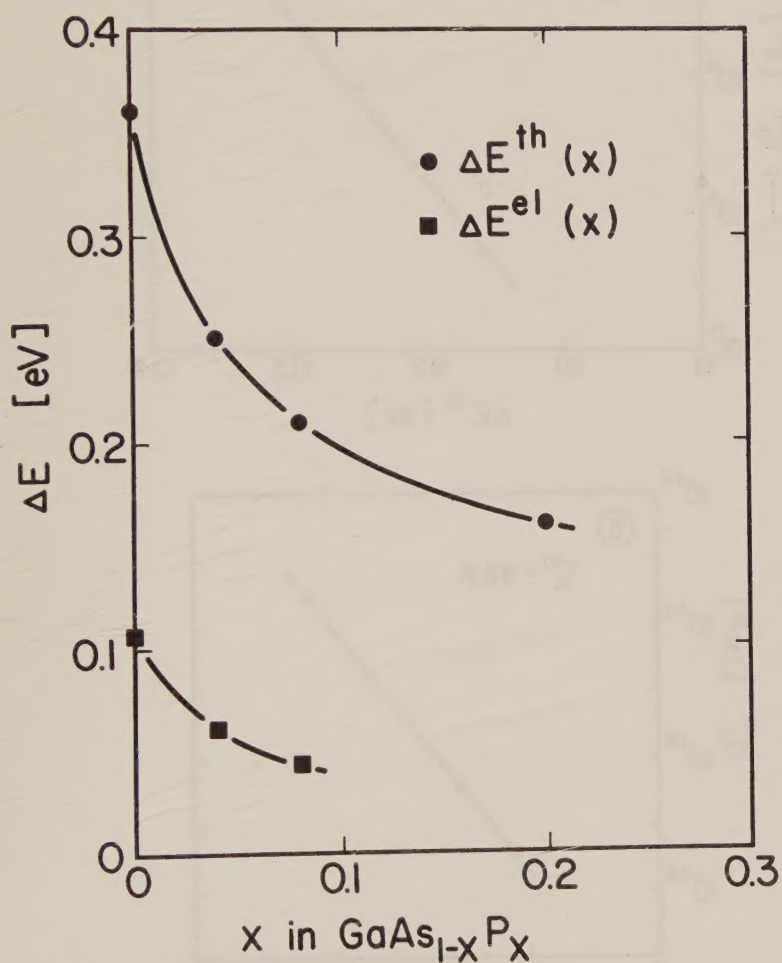


Fig. 7 The activation energies for thermal regeneration, ΔE^{th} , and electron induced regeneration, ΔE^{el} , plotted versus the alloy composition.

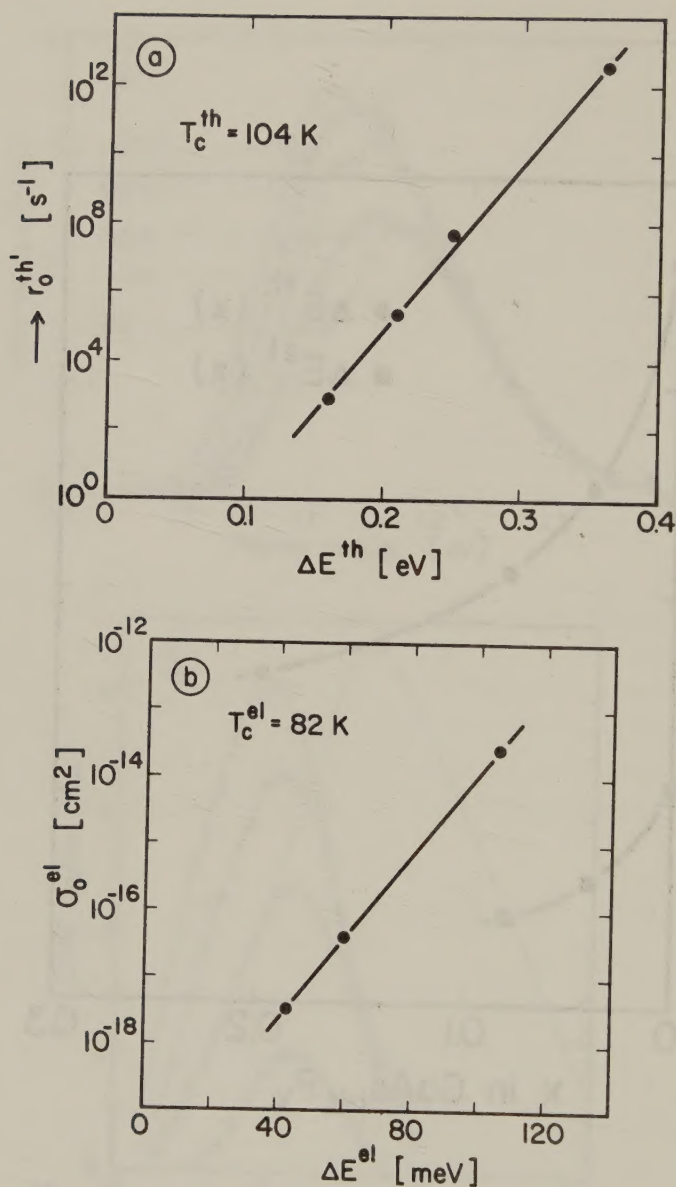


Fig. 8 a) The logarithm of the prefactor of the thermal regeneration rate, r_0^{th} , (see Eq. (3)) in the alloy system plotted as a function of ΔE^{th} .

b) The logarithm of the cross section for the electron induced regeneration rate, σ_0^{el} , (see Eq. (3)) in the alloy system plotted as a function of ΔE^{el} .

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